

Interplay between pairing and exchange in small metallic dots

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Abstract

We study the effects of the mesoscopic fluctuations on the competition between exchange and pairing interactions in ultrasmall metallic dots when the mean level spacing δ is comparable or larger than the BCS pairing energy Δ . Due to mesoscopic fluctuations, the probability to have a non-zero spin ground state may be non-vanishing and shows universal features related to both level statistics and interaction. Sample to sample fluctuations of the renormalized pairing are enlightened.

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The competition of superconductivity and ferromagnetism and their possible coexistence in bulk materials has been intensively investigated in the past and it lead to a very rich phase diagram [1]. In addition to the suppression of superconductivity due to time reversal symmetric breaking, new phases with non-uniform magnetic and superconducting ordering were predicted [2]. This picture does not hold when the size of the sample is reduced such that the average level spacing δ becomes comparable with the energy scales related to the onset of macroscopic order. In this case fluctuation (both thermal and quantum) effects become very important and even the definition of ordered phase is elusive. For instance a small superconductor will not posses a fully developed gap [3] and the spin of a ferromagnet will not be macroscopically large [4]. Nevertheless distinct features reminiscent of the macroscopic order can be traced even in nanosized grains.

In this Letter we investigate the competition between exchange and pairing in small metallic grains. Our approach starts from the results of a recent work of Kurland *et al.* [5] who have shown that universal features of electrons in isolated mesoscopic grains are accounted for by the Hamiltonian (see also Refs. [6, 7])

$$\mathcal{H} = \sum_{\alpha=1}^{\Omega} \sum_{\sigma=\{\uparrow,\downarrow\}} \epsilon_{\alpha} c_{\alpha,\sigma}^{\dagger} c_{\alpha,\sigma} + E_C \hat{N}^2 - \lambda \hat{T}^{\dagger} \hat{T} - J \hat{S}^2. \quad (1)$$

This description is valid in the limit of very large dimensionless conductance $g = E_T/\delta$, being E_T the Thouless energy and δ the mean level spacing. In Eq.(1) the first term is the kinetic energy while the electron-electron interaction is expressed as a sum of three terms, which describe respectively charging, pairing and exchange interactions. The index α spans a shell of Ω doubly degenerate ($\sigma = \pm$) time reversed single particle states of energy ϵ_{α} , and $c_{\alpha,\sigma}$ ($c_{\alpha,\sigma}^{\dagger}$) are the corresponding annihilation (creation) operators. The energies ϵ_{α} are distributed according to the Gaussian Orthogonal Ensemble (GOE), which describes the case of time-reversal symmetry with no spin-orbit coupling [8]. The interaction depends only on the collective variables $N = \sum_{\sigma} c_{\alpha,\sigma}^{\dagger} c_{\alpha,\sigma}$ (the number operator), $T = \sum_{\alpha=1}^{\Omega} c_{\alpha,-} c_{\alpha,+}$ (pair creation operator), and $\vec{S} = \sum_{\alpha=1}^{\Omega} c_{\alpha,\sigma}^{\dagger} \vec{\sigma}_{\sigma,\sigma'} c_{\alpha,\sigma'}$ (the total spin operator, σ are the Pauli matrices). For isolated grains N is fixed and the charging term can be ignored. Pairing interaction tends to favour the formation of spin singlets, while exchange tends to favour maximal spin, so they compete in determining the spin ordering.

A clear picture is already emerging in the case where either pairing or exchange is present. In the absence of exchange interactions ($J = 0$), the Hamiltonian in Eq.(1) describes pairing

correlations in small metallic grains [3]. Although no phase transition occurs, signatures of pairing correlations have been detected [9] even in nanosized grains. For this model it has been shown [10, 11] that the low-energy properties are universal functions of the ratio $\delta/\Delta = 2\sinh(\delta/\lambda)/\Omega$ (Δ is the BCS gap value). Upon increasing the size of the grains, hence decreasing the ratio δ/Δ , there is a crossover [11] between the case of ultrasmall grains ($\delta \gg \Delta$) where pairing produces strong quantum fluctuations [10], and a regime ($\delta \ll \Delta$), where the BCS mean-field description remains valid [12]. Signatures of pairing correlations may be detectable in thermodynamic quantities [13], even for ultrasmall grains. In absence of pairing ($\lambda = 0$) the properties of the model of Eq.(1) are determined by the interplay between the kinetic term, which favours Pauli filling of the levels and zero total spin in ground state, and exchange one which tends to maximize S and eventually leads to the Stoner instability for $J \geq \delta$. However in mesoscopic samples [5, 14, 15, 16] it is possible to find individual grains with a cluster of $2S$ closely spaced levels around the Fermi energy, whose ground state may have spin S even for $J \ll \delta$. The probability $P_S(J/\delta)$ of spin- S ground state directly reflects the universal properties of the level statistics [17].

In order to describe the interplay between superconductivity and ferromagnetism in small grains we study how the tendency to magnetic ordering is reduced when pairing is gradually increased. Hence, in the same spirit as in Ref. [17], we consider the probability P_S of finding a spin- S ground state in the regime $J, \lambda \ll \delta$, which is in turn related to the spontaneous magnetization of an ensemble of grains. All the results that will be presented are obtained in the half-filling scheme, where the numbers of electrons N is equal to the number of levels Ω . For an ensemble of normal grains $P_S(J/\delta)$ for $J \ll \delta$ is non-zero due to grains with a cluster of $2S$ close levels around the Fermi energy [17]. The same picture applies when pairing interaction is present except that, since the energy balance between spin S states involves the pairing correlation energy, one should take into account contributions coming from the entire shell of Ω levels. In the weak coupling limit $J, \lambda \ll \delta$ there is a simple way to circumvent this problem, namely to consider a shell of $2S$ levels with a renormalized pairing coupling $\tilde{\lambda}_{2S}$ (see Ref. [18]). Notice that whereas no sample to sample fluctuations affect the bare λ , the actual level distribution may produce fluctuations in $\tilde{\lambda}_{2S}$, which we ignore at the moment. With this hypothesis the probability to have spin- S in the ground state, for

$0 < J - J_S^* \ll \lambda \ll \delta$, is

$$P_S \left(\frac{J}{\delta} \right) = C_S \left(\frac{J}{\delta} \right)^{\alpha_S} \left(\frac{J - J_S^*}{\delta} \right)^{\alpha_S}, \quad (2)$$

where $\alpha_S = (S + 1)(2S - 1)/2$, J_S^* is the threshold value of the exchange below which the spin probability vanishes and depends on the spin and the pairing ($J_1^* = \tilde{\lambda}_2$, $J_{3/2}^* = 2\tilde{\lambda}_3/3$) and the C_S are dimensionless constant depending only on the spin S ($C_1 = \pi^2/3$, $C_{3/2} = 9\pi^4/50$) which directly reflect the universal statistical properties of the GOE level distribution. Compared to the case where pairing is absent [5], the exponent α_S is halved.

We now check these results with numerical calculations. We have considered grains with size up to $\Omega = 30, 31$, which are large enough to show the universal behavior of the pairing interaction [11], as in grains with much larger Ω . We considered ensembles of grains whose single particle spectra $\{\epsilon_\alpha, \alpha = 1, \dots, \Omega\}$ realize the GOE level statistics. To this end we diagonalized a set of $5\Omega \times 5\Omega$ real orthogonal random matrix taking out the central Ω eigenvalues, in order to avoid edge effects. Then we found the many-particle energies of Eq.(1) by using the Richardson exact solution [19] for larger systems ($\Omega \geq 20$), whereas for smaller systems we used the standard numerical diagonalization of the Hamiltonian or the Lanczos method. For systems with even (odd) N we studied the probability P_1 ($P_{3/2}$) of a ground state with spin 1 (3/2) using a set of 10^5 (10^6) level configurations (systems with odd N require more statistics because $P_{3/2}$ is smaller). Results are shown in Fig.1 and Fig.2 for values of J/δ such that the probability that the ground state has larger spin ($S > 1$ or $S > 3/2$) is negligible. The result given in Eq.(2) reproduces quite well the numerical data, except for smaller δ/Δ (large grains) and near J_S^* , a regime which we discuss later. Thus we conclude that P_1 and $P_{3/2}$ show the universal features of the level statistics predicted in Eq.(2), namely the coefficient C_S and the power law as a function of $(J - J_S^*)$. In addition we find a new manifestation of the universal behavior in the quantity J_S^* . According to the simple theory which leads to Eq.(2) for grains with even (odd) N this quantity is related to the renormalized two (three) levels pairing constant $\tilde{\lambda}_2$ ($\tilde{\lambda}_3$). By fitting the linear part of the curves in Fig.1 and Fig.2, we find that $\tilde{\lambda}_2$ and $\tilde{\lambda}_3$ are given by a universal functions of δ/Δ , of the form $\tilde{\lambda}_{2S} = \delta / \ln(a_S \delta / \Delta)$ [10, 11]. This result, shown in the insets of the Fig.1 and Fig.2, is valid over several decades of values of the parameters.

We now discuss more carefully the behavior near J_1^* for grains with even N (Fig.1). For samples with larger pairing interaction (smaller δ/Δ) the probability P_1 shows a non

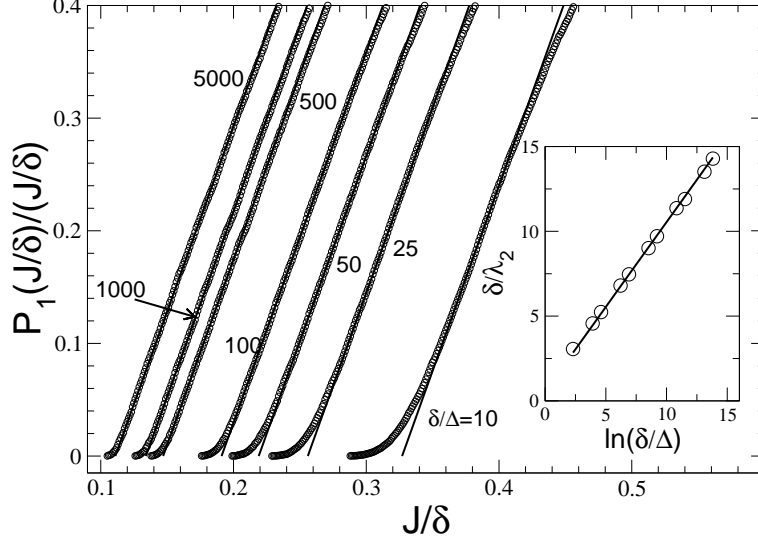


FIG. 1: The $P_1(J/\delta)$ probabilities in the case $\Omega = 30$ for the different values of the ratio δ/Δ (circles). The lines represent the fits to the data using the expression given in Eq.(2). In the inset, the numerical data of the renormalized pairing constant (circles) extracted from J_1^* are plotted as a function of δ/Δ . They coincide with the value of the renormalized pairing $\tilde{\lambda}_2 = \delta/\ln(a_1\delta/\Delta)$ (with $a_1 = 1.721$).

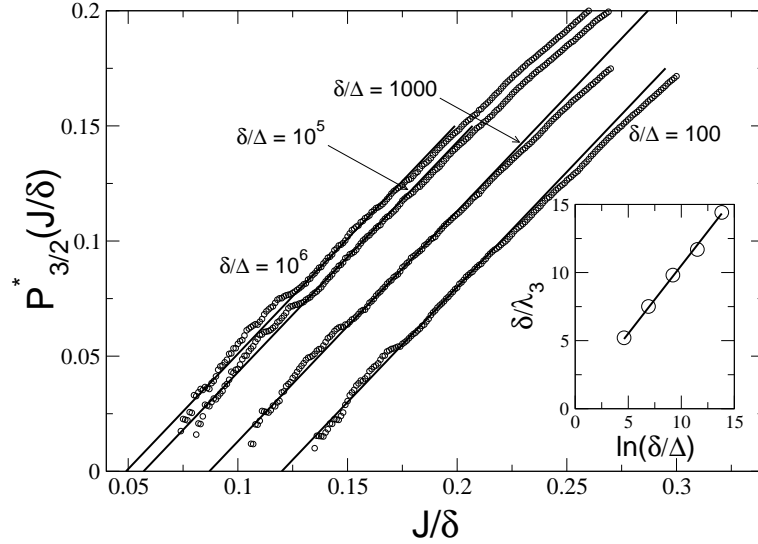


FIG. 2: The probabilities in the case $\Omega = 31$ for the different values of the ratio δ/Δ . For convenience we plotted $P_{3/2}^*(J/\delta) = [1350P_{3/2}(J/\delta)/\pi^4]^{(2/5)}/(9J/\delta) = (J - 2\tilde{\lambda}_3/3)/\delta$. The circles represent the numerical data, and the lines the linear fits. In the inset, the numerical data threshold value as a function of the ratio δ/Δ . The scaling behaviour is obtained as in the previous figure with $a_{3/2} = 1.679$.

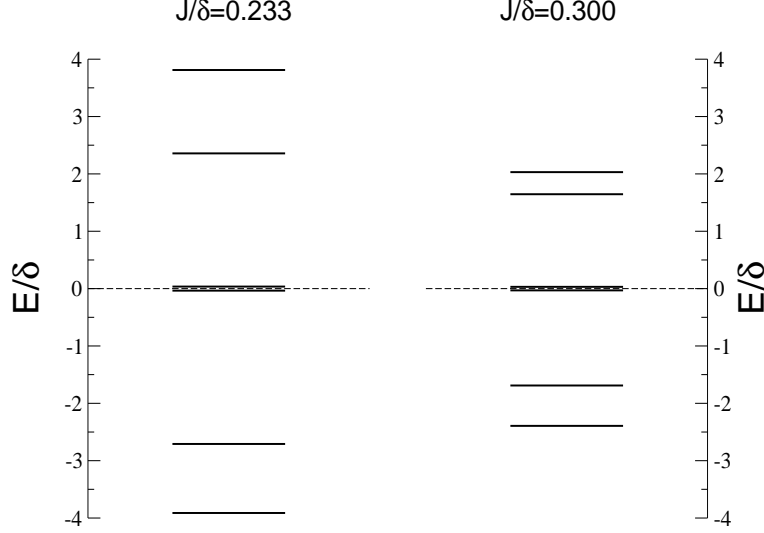


FIG. 3: A typical configuration of the single-particle energy levels (we chose as an example $\Omega = 6$ and $\delta/\Delta = 25$) which leads to a ground state with spin one in the tail region (left side, $J/\delta = 0.233$) and in the linear region (right side, $J/\delta = 0.300$). The Fermi level has been set to zero.

vanishing tail for $J \lesssim J_1^*$. We argue that this effect is due to sample to sample fluctuations of the *renormalized* $\tilde{\lambda}_{2S}$ which we ignored in Eq.(2). To understand this point consider even N and samples for which the two levels in the central cluster at the Fermi energy are closely spaced ($s_2 \ll \delta$). These samples may contribute to $P_1(J/\delta)$ for $J \sim J_1^*$. The two-level renormalized coupling in one of these grains is determined by the configuration of the other levels in the Ω shell and in particular depends strongly on the spacing s_4 between next neighboring pair of levels (above and below the Fermi energy, see the right side of Fig.3). For most of the samples $s_4 \approx 3\delta$ and they will have approximately the same two-level renormalized coupling, which is identified as the threshold J_1^* in Eq.(2). However for a small fraction of samples we may have $s_4 \gg 3\delta$ (see the left side of Fig.3) which leads to a smaller value of the renormalized coupling. As a consequence pairing correlations will be weaker and the ground state will have $S = 1$ for a value of J smaller than J_1^* . These samples determine the appearance of the tail for $J \lesssim J_1^*$.

To check this argument we have first analyzed the typical level configurations of samples contributing to $P_1(J/\delta)$ (Fig.3). Indeed samples which contribute to $P_1(J/\delta)$ in the tail region have level configurations as in Fig.3, with a central cluster of two levels around the Fermi energy, and all the others very far away, whereas in the region described by Eq.(2) the

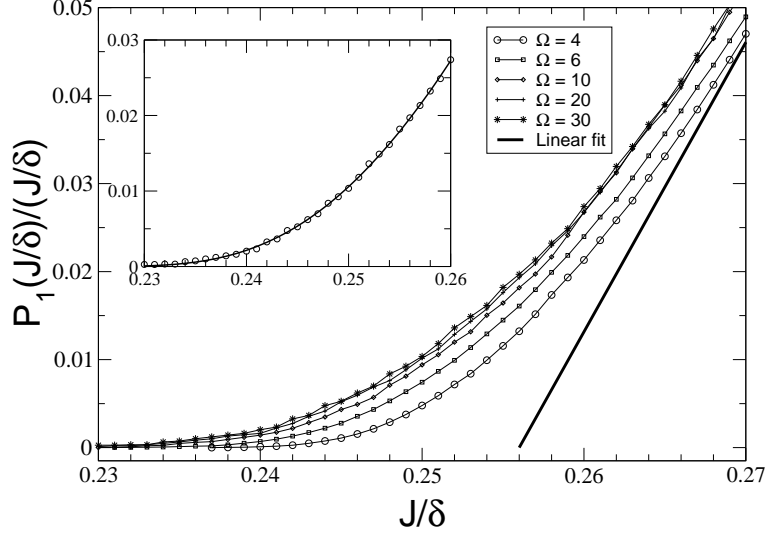


FIG. 4: The behavior of the spin-1 ground state probabilities for different values of the bandwidth Ω (curves with different symbols) at the fixed value $\delta/\Delta = 25$ in the tail region. The form $P_1(J/\delta) = (81/64\pi^2\delta^2)J(J - \lambda)\text{erfc}[b/(J - \lambda)]$ ($\text{erfc}(x)$ is the complementary error function) was chosen from the solution with three levels. The fit to the curve with $\Omega = 30$ (circles, in the inset) is obtained for $\lambda/\delta \sim \tilde{\lambda}_3/\delta = 0.201$ and $b = 6.821 \cdot 10^{-2}\delta$.

other levels are closer to the two central ones. Further insight can be gained by including in Eq.(2) the effect of fluctuations of a neighboring level. We consider a shell of three levels with two electrons. Two levels are close while the third lies far away. In this system we determine the approximate form of the probability distribution $P_1(J/\delta)$ (caption of Fig.4), which is non vanishing in the tail region. Moreover we fitted this result with numerical data for larger systems (up to $\Omega = 30$, see inset of Fig.4), using the value of the renormalized coupling as a fitting parameter(see the inset of the Fig.4). The agreement with the numerical data suggests that fluctuations of the renormalized pairing constant due to the statistics of far levels are negligible.

We also studied the behavior of $P_1(J/\delta)$ for $J \lesssim J_1^*$, for fixed $\delta/\Delta = 25$ and for various values of the bandwidth Ω . We used sets of GOE levels with 10^7 realizations for $\Omega = 4, 6, 10$ and with 10^6 realizations for $\Omega = 20, 30$. We see that for the values of J we could study $P_1(J/\delta)$ becomes independent on Ω for $\Omega \gtrsim 20$, showing that still $P_1(J/\delta)$ depends on features of the universal level statistics. However by decreasing J the behavior of $P_1(J/\delta)$ will depend more and more on details of the full Ω shell so universality is lost (for instance

$P_1(J/\delta)$ is expected to be nonzero for $J > \lambda$, the *bare* coupling). On the contrary by increasing J we recover the behaviour of Eq.(2) and all the curves collapse on the same line. The above analysis could be in principle carried out also in samples with odd N . However the unpaired electron in the spin 1/2 ground state weakens pairing correlations. As a consequence the tail in $P_{3/2}(J/\delta)$ is tiny and it would require a much larger statistics to be investigated.

Finally we consider grains with larger pairing interaction such that $\delta \lesssim \Delta$. This is the crossover region to BCS-like superconductivity [11, 18] and an analytical approach to the problem is a formidable task, so we studied this regime numerically. Some qualitative results can be inferred by looking at the evolution of the behavior of the probability distributions $P_S(J/\delta)$ from the perturbative region $\delta \gg \Delta$ to the region $\delta \sim \Delta$. In Fig.5 the probability distributions of the smallest and largest spin ground states are shown for the systems with an even ($\Omega = 30$) and odd ($\Omega = 29$) number of electrons in 10^5 GOE realizations. For smaller J we have $P_0(J/\delta) = 1$ whereas for large J the maximal spin is favoured. In the intermediate region the stable value of S is finite but the physics is sensitive to mesoscopic fluctuations. Results show that this crossover region becomes narrower as the ratio δ/Δ decreases. This trend is confirmed by the fact that, on increasing the exchange, the system tends to maximize the spin in the ground state, whereas in the dot regime $\delta > \Delta$ the crossover occurs gradually across all the increasing values of the spin. In the samples with an odd number of electrons the width of the J range between the probability of the smallest and largest spin probability is larger than in the even systems since the pairing is weakened by the presence of the unpaired electron.

In conclusion we studied a model for the competition between superconducting and ferromagnetic ordering in small metal grains. The presence of pairing implies that all the levels in the shell are responsible for the determination of the spin in the ground state. As compared to the normal case there are three new features: i) a soft threshold appears in the P_S directly related to the renormalized pairing coupling; ii) the power law behavior has a new exponent as compared to the case where pairing is absent; iii) at low enough exchange couplings $P_S(J/\delta)$ is determined by sample to sample fluctuations in the pairing coupling. By increasing both the pairing interaction and the exchange coupling we found that the region of the phase diagram where mesoscopic effects are important becomes progressively narrower.

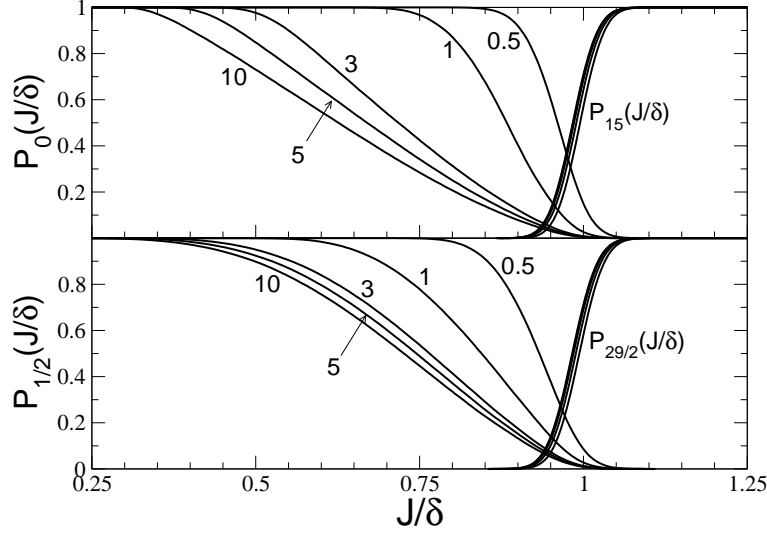


FIG. 5: The behavior of the probability distributions in the systems with an even (upper graph, $\Omega = 30$) and odd (lower graph, $\Omega = 29$) number of electron, of the smallest ($S = 0$ and $S = 1/2$, decreasing curves on the left side) and largest ($S = 15$ and $S = 29/2$, curve on right side of the figure) spin ground states for different values of the δ/Δ ratio (denoted by the numbers near each curve, in the case of largest spin value all the curves collapse on a single one) in a set of 10^5 realizations.

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